

### Description of the Invention

Applicant's invention relates to a method for preparing membrane electrode assemblies (MEAs), and in particular to a method of manufacturing a proton-conducting cation-exchange electrolyte membrane for use in a membrane electrode assembly (MEA), in which atmospheric pressure plasma deposition is used to deposit catalysts such as platinum onto a polymer substrate, or a substrate including carbon cloth or carbon particles. The invention has three principal characteristics:

- 1) The noble metal catalyst is deposited on the membrane by discharge enhanced chemical vapor deposition (DECVD); and
- 2) The DECVD is carried out at atmospheric pressure, without adding noble gases to the single mixed randomized DECVD carrier gas/reactant stream which is the only gas stream in the process where the single mixed randomized gas stream of reactants in a carrier gas is the only gas stream which passes directly between said 2 or more electrodes.
- 3) The reactants included in a carrier gas pass directly between 2 or more electrodes

### REMARKS

Claims 1, 3-9, and 11-16 are pending, and stand rejected.

Claim 1 has been amended to cite "wherein said single mixed randomized gas stream of reactants in a carrier gas is the only gas stream which passes directly between said 2 or more electrodes.". This amendment is supported by the Diagrams, in which there is only a single inlet 1 in which the reactants are introduced – and also supported by the description in paragraph [0022], first sentence.

### Response to the Examiner's Response to Applicant's Arguments:

#### **Fukuda:**

The Examiner contends that Applicant's "single mixed randomized gas stream" does not necessarily mean "single" or "mixed" or "randomized". Applicant has amended

claim 1 to now say the same thing in another form, to remove any doubt that one may have had as to the clear meaning of the claimed invention.

“Single”: The Examiner contends that “single” does not necessarily mean “one”, since “additional non-mixed gas streams are not excluded from the claim”. Applicant believes that a “single” gas stream would mean to one of ordinary skill in the art the same as Webster’s Collegiate Dictionary definition of “single” as “unaccompanied by others; lone, sole”. Applicant has amended claim 1 to state in other terms that the claimed single gas stream is the only gas stream that passes directly between the electrodes, making it even more clear that the additional non-mixed gas streams of the Fukuda reference would be excluded from Applicant’s claims.

Applicant respectfully believes that the Examiner’s interpretation of the “comprising” language of claim 1, which allows for other process steps and parameters, is overly broad. “Comprising” does allow for added steps and parameters. Applicant’s claim cites “...comprising the step of...”, and Applicant agrees that other steps can occur before and after the claimed step. However, Applicant believes that “...comprising the step of...” does not open the description of that particular step to wild inclusion of things not mentioned or suggested in that particular step. Since the claimed step requires a “single” gas stream, other gas streams are not allowed, and one of skill in the art would not interpret the term “single” in the context of this claim to mean more than one.

“Mixed” and “randomized”: These two terms are related in Applicant’s claim. Webster’s Collegiate Dictionary defines “randomize” as arranged in a random way, with “random” meaning “lacking a definite pattern”. Applicant believes that a person of ordinary skill in the art would understand “mixed randomized” to mean a single gas stream in which the reactants and carrier gas are mixed in a near homogenous manner. The Examiner reads “mixed randomized” to mean the Fukuda at least three different unique separate compositions – 1) an outer inert gas; 2) an inner reactant gas; and 3) a finite interface between the two gas streams in which some mixing of the two stream occurs. The Fukuda reference specifically designs to minimize any interfacial mixing, so the gas stream consists primarily of two separate gas streams.

Applicant contends that the Fukuda three-strata gas streams do not teach or suggest Applicant’s claimed single mixed randomized gas stream. Nor would one of

ordinary skill in the art arrive at Applicant's single mixed randomized gas stream based on Fukuda's teaching of a three-strata gas streams. Moreover the three-strata gas streams of Fukuda teach away from Applicant's claimed single mixed randomized gas stream.

The Examiner maintains that "the process of Fukuda '306 will necessarily a degree of mixing at the interface between the two gases, reactant and carrier". (Page 2, 8/21/07 office action.) Applicant totally agrees with the Examiner, and it is this exact point that differentiates Applicant's claim from the Fukuda reference! While there is some minimal mixing of the gas streams at the interface, there is clearly no single randomized gas stream. The Fukuda reference apparatus design specifically attempts to minimize any turbulent flow and any mixing of the gases. Therefore, as the Examiner has pointed out, there is an interface with minimal mixing, but the mere presence of any "interface" is a clear indication of two separate phases, perhaps with a small gradient. However, it is clear that the gas phases of Fukuda are meant to remain separate, and clearly not randomized as in Applicant's claims.

The Fukuda reference clearly teaches that "the apparatus is constructed so that the reactive gas is not directly in contact with the discharge surface of the first electrode or the second electrode" (Col 3, lines 28-31). In column 15, lines 29-45 the Fukuda reference describes the workings diagramed in Figure 2. "The voltage application electrode is provided so that the electrode is surrounded with the gas paths, whereby turbulent flow is difficult to occur in the discharging space, the inert gas contacts the voltage application electrode 2, and the reactive gas contacts the surface of substrate 1. The above structure of the electrode section is such that the voltage application electrode 2 does not directly contact the reactive gas for forming a layer..."

Thus, while the Examiner may conclude that some small amount of mixing of the two gas streams may occur, it is clear from the Fukuda reference that the apparatus is designed to either totally prevent any mixing of the two gas streams (as in Figure 1), or else extreme pains are taken to minimize any mixing by limiting turbulent flow.

Meanwhile, Applicant's invention involves "the vaporized reactants and carrier gases are directed through the discharge" (page 12, para. [0025], lines 2 and 3). Applicant's do not "use a noble gas in the carrier gas to suppress arcing...there is

adequate randomization of microarc location and/or downstream gas mixing.” (page 14, [0029])

The Fukuda teaching of minimizing or avoiding contact and mixing of the reactive and inert gases not only fails to teach or suggest Applicant’s claim limitation of a randomized gas stream, but clearly teach away from Applicant’s claims. One of skill in the art would not be motivated by the “separate stream” teaching of Fukuda, to practice Applicant’s claims of a “single mixed randomized gas stream”.

**Hammerschmidt in view Fukuda and JP 10-275698**

Claims 1, 3, 5-6, and 11-14, stand rejected under 35 U.S.C. 103(a) as being unpatentable over Hammerschmidt (US 6,010,798) in view of Fukuda (US 6,849,306), Fukuda US 2003/02032136, and JP 10-275698. These references fail to create a *prima facie* case of obviousness over Applicant’s claims, as amended.

The Hammerschmidt reference describes a novel polymer electrolyte membrane arrangement. It offers little insight into the method for producing a proton-conducting cation-exchange electrolyte membrane, which is the subject of Applicant’s invention. The Hammerschmidt reference describes only generally the process for chemical deposition as a “plasma-chemical process”, using as examples chemical deposition at “low-pressure plasma between  $10^{-4}$  and 10 mbar”, and as an alternative “sputtering methods”. (Col 3, lines 40 – 55). Both of these methods are very different from the atmospheric pressure plasma deposition method claimed by Applicant, and the differences have been clearly discussed in the many responses to rejections filed by Applicant in this application.

The Hammerschmidt reference fails to teach or suggest a discharge enhanced chemical vapor deposition process carried out at atmospheric pressure without adding a noble gas and having a single mixed randomized gas stream of reactants in a carrier gas which is the only gas stream passing directly between 2 or more electrodes, and therefore fails to present a *prima facie case* of obviousness. One of ordinary skill in the art would not be motivated by a teaching of a membrane, and disclosure only of methods for forming that membrane that do not contain all of Applicants claimed elements and limitations, to practice Applicant’s claims.

The Fukuda references have been discussed above. The Fukuda references also fails to teach or suggest a discharge enhanced chemical vapor deposition process carried out at atmospheric pressure without adding a noble gas and having a single mixed randomized gas stream of reactants in a carrier gas which is the only gas stream passing directly between 2 or more electrodes, and therefore fails to heal the deficiencies of the Hammerschmidt reference in providing a prima facie case of obviousness when combined with that reference.

JP 10-275698 is cited as a secondary reference to show that atmospheric plasma can be formed without a rare gas. The JP 10-275698 solution is to begin with a mixture of oxygen and helium, then turn off the helium and continue plasma generation to perform ashing or etching. The JP'698 reference makes no mention of a single mixed randomized gas stream of reactants in a carrier gas, and therefore fails to heal the deficiencies of the other cited references to present a prima facie case of obviousness.

Hammerschmidt in view of Fukuda, JP 10-275698 and Others (generally):

As shown above, the Hammerschmidt reference in view of Fukuda fails to teach or suggest all of Applicant's claim limitations, in particular: A catalyst deposited on a membrane by discharge enhanced chemical vapor deposition (DECVD); at atmospheric pressure, without adding noble gases to the single mixed DECVD carrier gas/reactant stream, where the reactants included in a carrier gas pass directly between 2 or more electrodes. The Examiner then cited additional references, none of which, taken with Hammerschmidt and Fukuda, cite all of Applicant's claim limitations (as amended). Therefore, no *prima facie* case of obviousness is presented.

Hammerschmidt in view of Fukuda, JP 10-275698 and Schutze

Claim 4 stands rejected under 35 U.S.C. 103(a) as being unpatentable over Hammerschmidt (US 6,010,798) in view of Fukuda (US 6,849,306), Fukuda US 2003/02032136, and JP 10-275698, further in view of Schutze. The Schutze reference teaches a plasma jet using flowing helium. Applicant's amended claims cite a method without adding noble gases to the DECVD carrier gas. The Schutze reference

not only fails to teach or suggest Applicant's claim limitation of no added noble gas, but teaches away from Applicant's claims by requiring a noble gas. The Schutze reference fails to correct the deficiencies of the other cited references, fails to teach or suggest Applicant's claim limitations, and teaches away from Applicant's claims.

#### Further in view of Yasumoto

Claim 7 stands rejected further in view of Yasumoto (US 2003/0096154). The Yasumoto is a secondary reference cited by the Examiner to teach the spraying of the catalyst onto the surface of the polymer electrode membrane. Applicant's do not claim a method in which a catalyst is sprayed onto a polymer electrode membrane, but rather a discharge enhanced chemical vapor deposition method. Thus the Yasumoto reference fails to teach Applicant's claims.

#### Further in view of Nanaumi

Claims 8-9 stand rejected under 35 U.S.C. 103(a) as being unpatentable further in view of Nanaumi (US 2004/0180250).

The Nanaumi reference is cited to cite polymer electrolyte membrane structures. However the Nanaumi reference fails to teach or suggest Applicant's many claim limitations, and fails to correct the many deficiencies of the other references cited.

#### Further in view of Kamo

Claims 14 and 15 stand rejected under 35 U.S.C. 103(a) as being unpatentable over Dearnaley (US Patent Number 6,159,533) in view of Schutze in view of Fornsel (WO 01/32949, US 6,800,336), and further in view of Kamo (US 2003/0059659). The Kamo reference is a secondary reference cited to show the use of a platinum alloy in the anode side of an electrolyte membrane. While the Kamo reference discloses a platinum/ruthenium alloy for a fuel cell electrode, the platinum/ruthenium alloy is supported on a carbon powder, rather than directly on a membrane as claimed by Applicant. In Example 2, the platinum/ruthenium alloy is screen printed using a slurry.

One in the art would not be motivated by this method alone – or in combination with the other cited reference to practice all of the limitations in Applicant's amended claims.

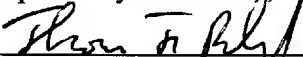
Further in view of Haug

Claim 16 stands rejected under 35 U.S.C. 103(a) as being unpatentable over Dearnaley (US Patent Number 6,159,533) in view of Schutze in view of Fornsel (WO 01/32949, US 6,800,336), and further in view of Haug. The Haug reference is a secondary reference cited to show the deposition of multiple catalyst layers. The Haug reference demonstrates the use of a vacuum sputter deposition system for producing a PEM. The disclosure of a multiple layer of catalyst by methods teaching away from Applicant's claimed method fails to heal the defects of the cited art to present a *prima facie* case of obviousness.

Conclusion

The references cited, either alone or in combination, fail to teach or suggest all of Applicant's claim limitations, and therefore fail to present a *prima facie* case of obviousness over Applicant's amended claims. For the above reasons the present claims 1, 3-9, and 11-16 are believed by the Applicant to be novel and unobvious over the prior art, thus the claims herein should be allowable to the Applicant. Accordingly, reconsideration and allowance are requested.

Respectfully submitted,

  
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